#### Status of LOCA Research in the High Burnup Cladding Performance Program\*

M.C. Billone, R. V. Strain, and Y. Yan

Argonne National Laboratory Energy Technology Division Argonne, IL 60439

The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory ("Argonne") under Contract No. W-31-109-ENG-38 with the U.S. Department of Energy. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

February 2002

<sup>\*</sup>Work supported by the U.S. Nuclear Regulator Commission, Office of Nuclear Regulatory Research. To be presented at the Fuel Safety Research Specialists' Meeting, Tokai, Japan, March 4-5, 2002

#### STATUS OF LOCA RESEARCH IN THE HIGH BURNUP CLADDING PERFORMANCE PROGRAM

M. C. Billone, R. V. Strain and Y. Yan Argonne National Laboratory (ANL) Argonne, IL USA

#### **ABSTRACT**

The High Burnup Cladding Performance program is being conducted at ANL to provide data in support of Loss-of-Coolant Accident (LOCA) and Reactivity-Initiated Accident (RIA) licensing criteria assessments for fuels at high burnup, as well as licensing criteria assessment for spent nuclear fuels under dry storage conditions. The USNRC Office of Nuclear Regulatory Research and the Electric Power Research Institute (EPRI) sponsor the program. Seven Limerick (GE-11 design) BWR fuel rods (≈57 GWd/MTU), ten H. B. Robinson (15x15 design) PWR fuel rods (≈67 GWd/MTU) and two TMI-1 (15x15 design) PWR fuel rods (≈49 GWd/MTU) have been provided to ANL for testing purposes.

The current LOCA acceptance criteria (10 CFR50.46) limit peak cladding temperature to 2200°F (1204°C) and maximum Equivalent Cladding Reacted (ECR) to 17% during high temperature steam oxidation to ensure adequate ductility during the Emergency Core Cooling System (ECCS) quench and during possible post-LOCA seismic events. In addition, NRC Information Notice 98-29 specifies that the ECR should be based on the total oxidation, including oxide layers formed during normal reactor operation. For high burnup PWR cladding, this would leave very little margin for the LOCA transient oxidation. The primary high burnup phenomena that may affect cladding response during ballooning and burst, steam oxidation, quench and post-quench events are: loss of base metal thickness due to oxidation, hydrogen pickup and formation of an inner-surface oxide layer, all during normal operation; the effective thickness and chemistry (i.e.,  $H_2$  and  $O_2$  content) of the prior-beta phase layer following steam oxidation and quench; decreased fuel permeability to gas flow from the plenum; and the tightness of the fuel-cladding bond. The LOCA Integral Tests will be conducted with high burnup fueled cladding segments (300-mm long) in order to include all the phenomena highlighted. The oxidation kinetics results at  $\approx 1200$ °C are being used to plan the LOCA Integral Test experimental times that correspond to ECR values equal to 17% and above and below 17%.

The first LOCA Integral Tests will be conducted using fueled Zircaloy-2 (Zry-2) samples from a Limerick BWR rod. Progress made to date includes: determination of the oxidation kinetics of archival and irradiated Limerick Zry-2 (1000-1200°C); construction/testing of an out-of-cell, "hands-on" LOCA mockup apparatus; detailed design for the remotely-operated in-cell LOCA apparatus based on the mockup experience; construction of, and out-of-cell testing in, the LOCA Integral Test apparatus; and construction of devices to partially remove fuel from the sample top/bottom and to weld end-caps and thermocouples to the sample, as well as to perform overall test train assembly. The BWR LOCA tests will be conducted with pre-stabilization at 300°C, 5 °C/s temperature ramp, hold time at 1204°C based on desired ECR, slow (≈3°C/s) furnace cooling to 800°C, and water quenching initiated at 800°C. For the first test, "plenum" volume, sample internal pressure and test time have been set at 10 cm³, 8.7 MPa and 10 minutes, respectively. Based on the oxidation kinetics tests, Cathcart-Pawel model predictions agree quite well with weight-gain and oxide-layer-growth data for unirradiated and irradiated Zry-2. However, for irradiated Zry-2, enhanced (≈46%) alpha-layer growth during oxidation and formation of oxygen-stabilized alpha "islands" within the prior beta layer during cooling lead to a smaller effective ductile layer, which may have a negative impact on the quench and post-quench ECR failure thresholds.

#### STATUS OF LOCA RESEARCH IN THE HIGH BURNUP CLADDING PERFORMANCE PROGRAM

M. C. Billone, R. V. Strain and Y. Yan Argonne National Laboratory, USA

#### Fuel Safety Research Specialists' Meeting

Session 3: LOCA-related Research on High Burnup Fuel JAERI Advance Science-Research Center Tokai, Japan March 4-5, 2002

# ANL LOCA-RELEVANT TESTS FOR HIGH BURNUP FUEL CLADDING

#### Steam Oxidation Kinetics Studies

- 900-1300°C, emphasis on 1204°C for 5-20 minutes
- Emphasis on kinetics of weight gain, (oxide + ") layer growth rate, effective \$ layer thickness vs. ECR

#### • LOCA Integral Tests

- Test adequacy of 10CFR50.46 ECCS licensing criteria (ECR  $\Omega$  17%, T  $\Omega$  1204°C) for high burnup fuel
- Determine high burnup fuel failure threshold for thermal quench fragmentation
- Post-Quench Ductility Tests (e.g., Ring Compression)

#### HIGH BURNUP LOCA ISSUES

- BWR Fuel Rods (Limerick at ≈57 GWd/MTU, ≈10-: m OD Oxide)
  - Effect of tight fuel-cladding bond and restricted gas flow on ballooning, burst, inner-surface-oxidation/hydrogen-pickup, etc.
  - Effect of irradiation on high temperature oxidation in steam
  - Effect of fuel-cladding mechanical interaction on fragmentation resistance during water quench; post-quench ductility
- PWR Fuel Rods (HBR at ≈67 GWd/MTU, Ω100-: m OD Oxide)
  - Similar fuel-cladding issues as for BWR
  - Effect of in-reactor oxide layer on oxidation kinetics and ECR.
  - Effect of hydrogen pickup on oxidation kinetics, fragmentationresistance during water quench and post-quench ductility.

#### • ECCS Acceptance Criteria

- Are ECR ≤ 17% and PCT ≤ 2200°F (1204°C) conservative?
- Margins to quench fragmentation and post-quench embrittlement?
- Include pre-transient oxidation in ECR calculation?

#### LOCA INTEGRAL TESTING SCOPE

#### Parameters Common to BWR and PWR Tests

- Fuel-cladding samples = 305-mm long; fueled region = 270 mm
- PCT =  $1204 \overline{20}$ °C, temperature ramps relevant to SB-LB LOCA
- Internal pressure  $P_i < 1.3 \times \text{system}$  pressure, plenum V = 5 to 10 cc
- Best-estimate 17% Ω ECR < ≈30% ≡ oxidation time ≈2-20 min.

#### High Burnup BWR Rods (Limerick)

- Temperature ramp rate =  $5^{\circ}$ C/s (2.5- $7^{\circ}$ C/s for SB-to-LB LOCA)
- Cladding)  $P = P_i P_s \Omega 8.6 \text{ MPa } [6.7 \text{ MPa } (\text{SB}) 8.6 \text{ MPa } (\text{LB})]$

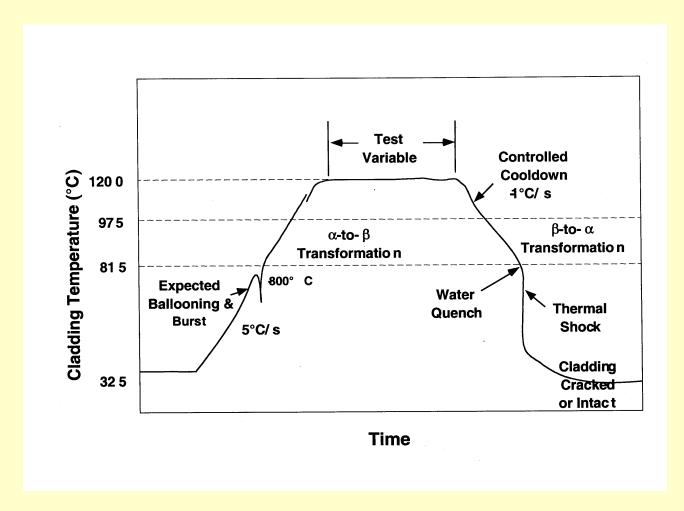
#### • High Burnup PWR Rods (H. B. Robinson)

- Temperature ramp rate =  $5^{\circ}$ C/s (1- $2^{\circ}$ C/s for SB, 7- $10^{\circ}$ C/s for LB)
- Cladding)  $P = P_i P_s < 20 \text{ MPa} [P_s = 3.4 \equiv 0.2 \text{ MPa} (SB \equiv LB)]$

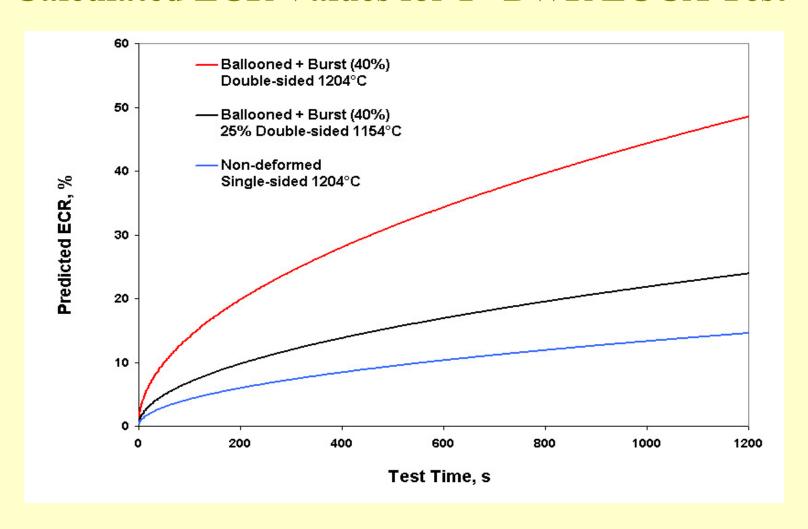
#### LOCA INTEGRAL TESTING SCOPE

- Steam and Quench Water Flow-rates/Volumes
  - Steam flow = 2 g/minute
  - Cool-down rate =  $3^{\circ}$ C/s from  $1204^{\circ}$ C to  $800^{\circ}$ C (1- $8^{\circ}$ C/s for BWR)
  - Quench water velocity = 5 mm/s (initiated at 800°C)
- First BWR Test Time = 10 min. = ?? Measured ECR
  - Ignore in-reactor-formed oxygen pickup (10 [m) in ECR
  - Wall thickness before transient  $\approx$ 0.7 mm (0.71 mm as-fabricated)
  - Use Cathcart-Pawel (CP) best-estimate oxidation model
  - Assume double-sided oxidation around 25% of circumference
  - Assume wall-thickness reduction  $\Omega$  40% in burst region
  - ECR = 10% for 1-sided oxidation, non-ballooned region at 1204°C

#### CLADDING PEAK TEMPERATURE RAMPS BASED ON PWR LOCA (PUMP-DISCHARGE LEG BREAK)



#### Calculated ECR Values for 1st BWR LOCA Test



#### LOCA INTEGRAL TEST APPARATUS

#### Out-of-Cell LOCA Apparatus

LOCA Mock-up

Samples: Zircaloy-2 tubes with zirconia pellets

Component development and checkout

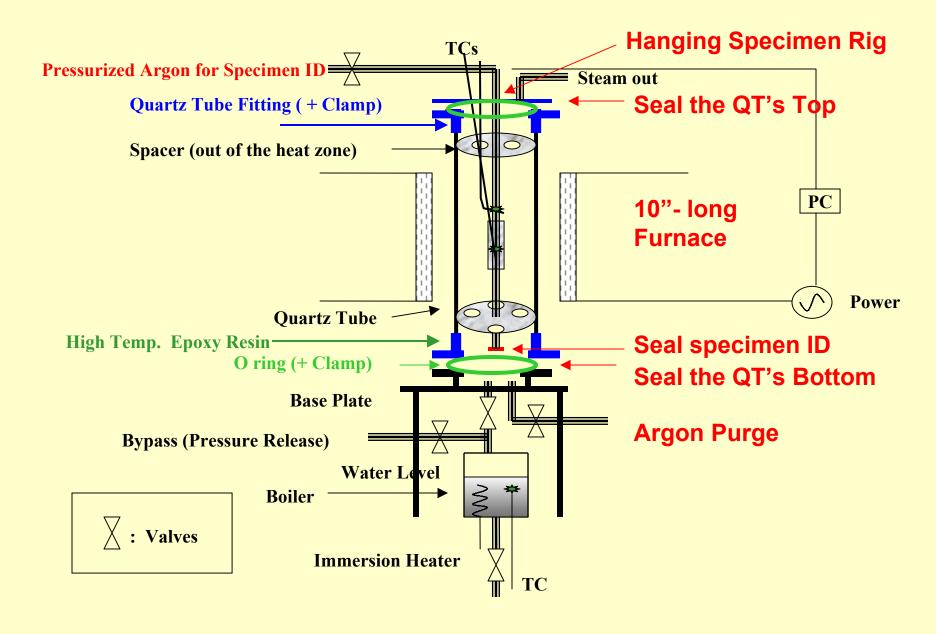
Experience used to design/construct final in-cell apparatus

Identical system to in-cell apparatus will be maintained out-of-cell;
 allows hands-on work to be performed in parallel with in-cell tests

#### In-cell LOCA Apparatus

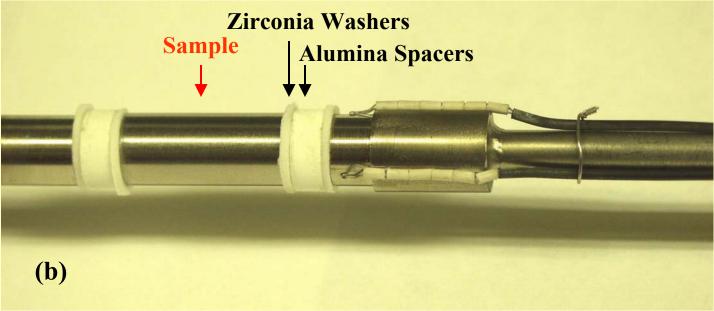
- Capable of doing both oxidation and LOCA tests
- Oxidation part of LOCA apparatus is operable in-cell
- In-cell installation of the internal pressurization , quench, etc. will commence following out-of-cell benchmark tests
- Conduct 1st Limerick BWR LOCA Test in April 2002

#### Schematic illustration of the new oxidation system



#### **Test Rig for Improved Oxidation Apparatus**





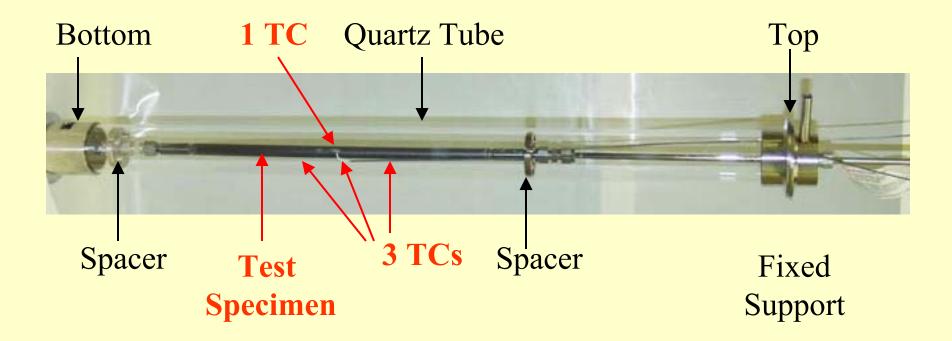
# 8.38 0.74 12.00 16.20

# LOCA Hanging Test Train

(Units in inches)



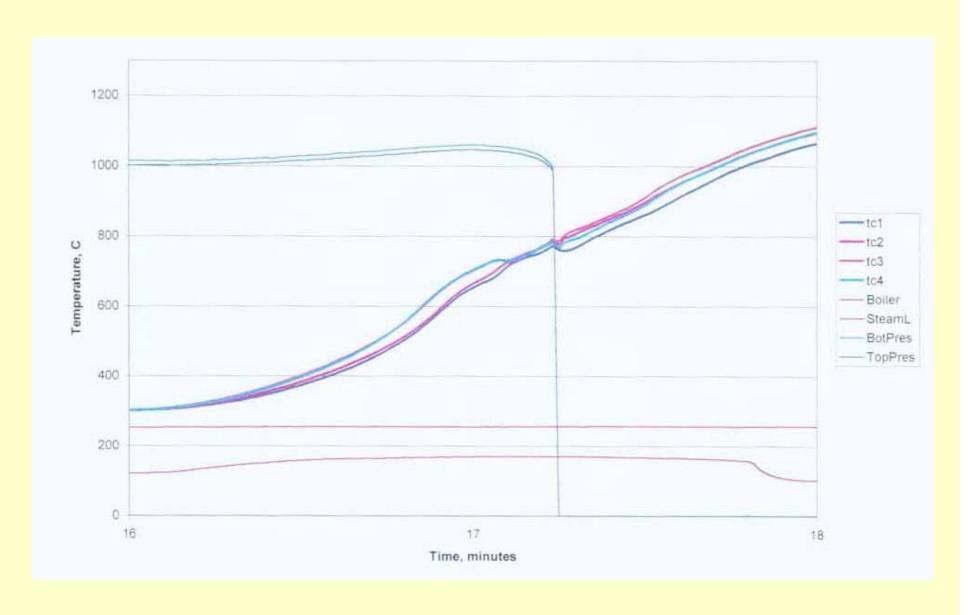
#### LOCA TEST TRAIN ASSEMBLY



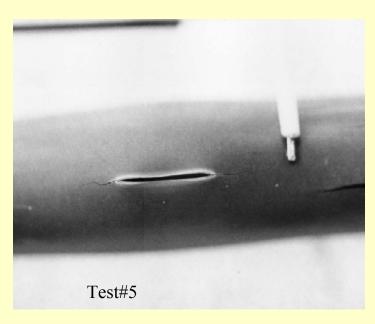
#### **SUMMARY OF LOCA MOCK-UP TESTS**

- Tests Conducted with GE-10 Zry-2 Cladding (0.81-mm wall) with Zirconia Pellets and void (gas) volume of only 3.6 cm<sup>3</sup>
- Unpressurized Test (#6) to Benchmark Temperature Distribution  $(T_{\Theta})$  at Specimen Midplane and Oxidation Kinetics
  - $T_{\theta} = 1211 \pm 15^{\circ}C$  (based on 4 TCs 90° apart)
  - Weight gains and oxide layer thickness determined from metallography and oxygen measurements agree with Cathcart-Pawel model predictions
- Pressurized Tests (#5, #7, #8, #9) with water quench
  - Internal pressure set at ≈7 MPa, remains relatively constant during ramp
  - $T_{\theta} = 1208 \pm 15$ °C for Test #8 (based on 4 TCs 90° apart)
  - Characterization of ballooned/burst region:
    - $\Delta D/Do \approx 40\%$
    - Burst at  $\approx$ 760-800°C, small burst openings  $\approx$ 10-mm by  $\approx$ 1-mm
    - Oxidation of <25% of inner surface

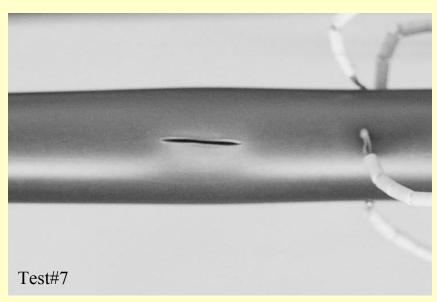
#### **CONDITIONS AT CLAD BURST - TEST #8**

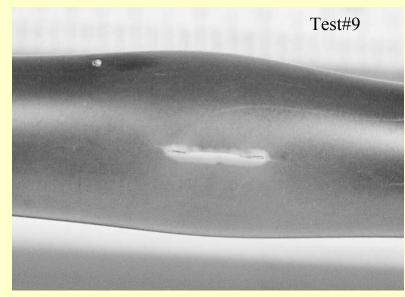


#### PHOTOS OF BREACH SITES

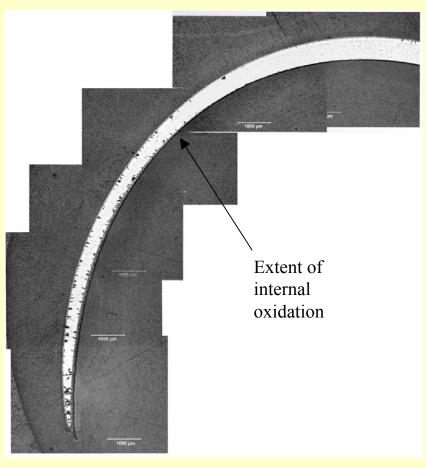


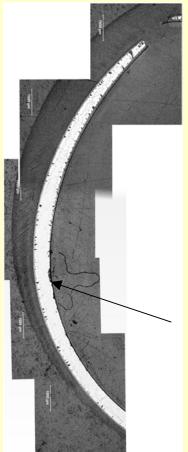






# Breach Site, Test #8 Transverse Section





Extent of internal oxidation

#### **OXIDATION KINETICS RESULTS (= 1200°C)**

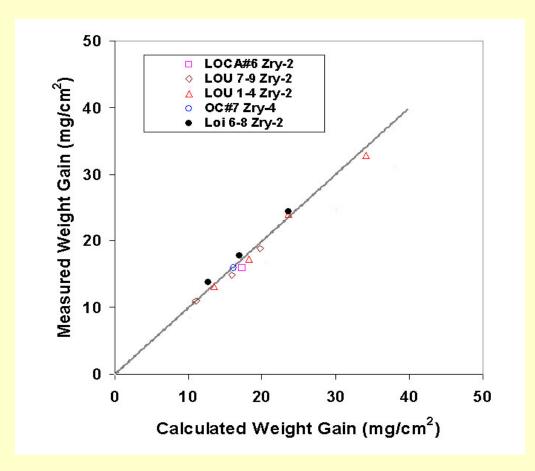
#### • Tests Conducted In-Cell for 5, 10 and 20 Minutes

- Unirradiated (archival) Limerick Zircaloy-2 (Zry-2)
- High burnup Limerick Zry-2 (=10-μm OD oxide, =70 wppm H)
- PWR Zry-4 (=49 GWd/MTU, =30-μm OD oxide, =150 wppm H)

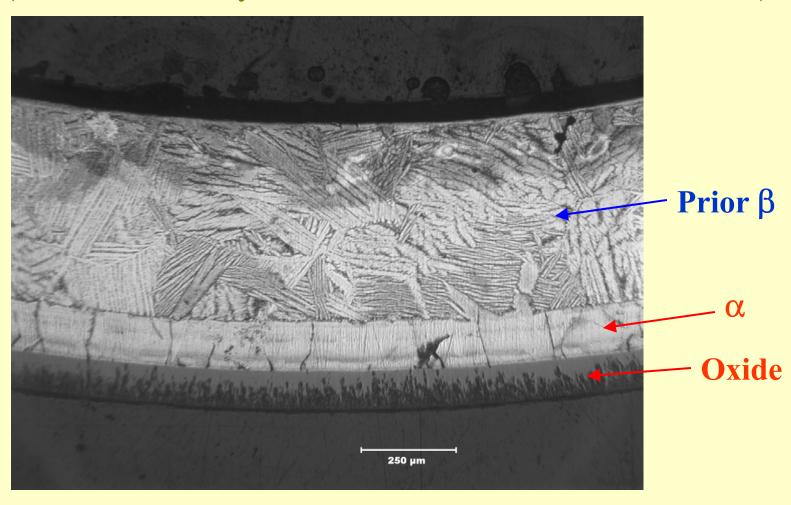
#### Results to Date (Detailed MET Analysis in Progress)

- Weight gain based on MET analysis of oxide,  $\alpha$  and  $\beta$  layers most reliable (vs. direct sample weight and oxygen increases)
- Weight gain kinetics data agree (within 6%) with Cathcart-Pawel (CP) best-estimate model predictions
- Enhanced growth of oxygen-stabilized  $\alpha$  layer observed
- Highly irregular  $\alpha$ - $\beta$  interface observed for irradiated cladding

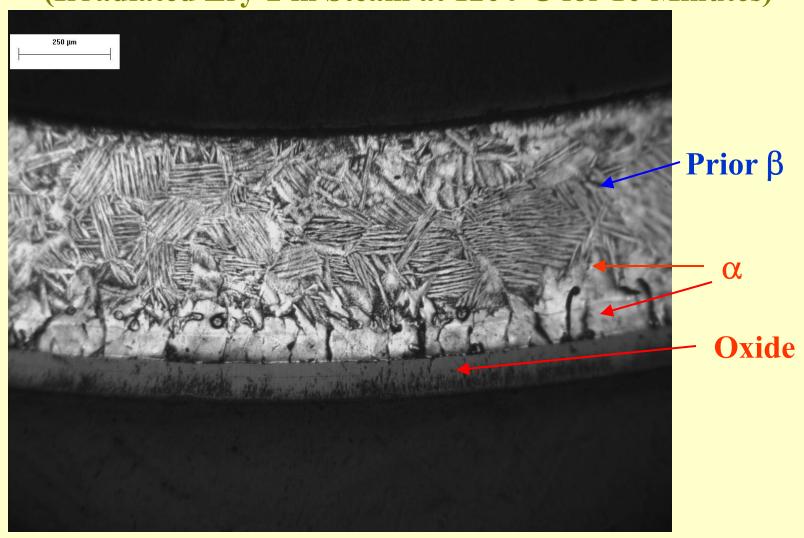
### WEIGHT GAIN FOR UNIRRADIATED AND IRRADIATED (LOI 6-8) LIMERICK ZRY-2



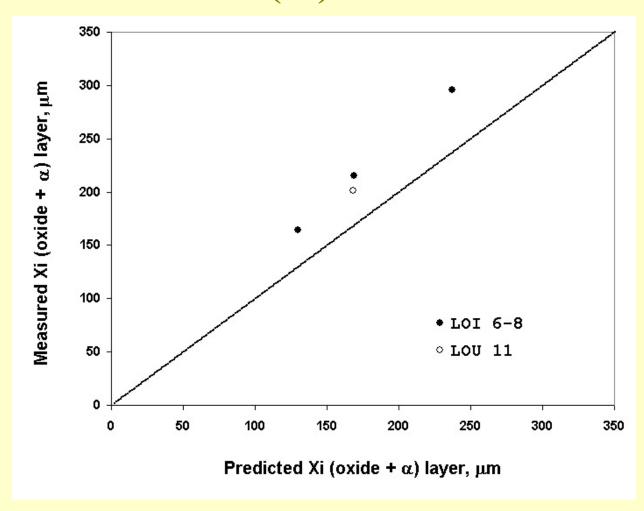
## OXIDE, $\alpha$ AND $\beta$ LAYER CHARACTERISTICS (Unirradiated Zry-2 in Steam at 1204°C for 10 Minutes)



## OXIDE, $\alpha$ AND $\beta$ LAYER CHARACTERISTICS (Irradiated Zry-2 in Steam at 1204°C for 10 Minutes)



# OXYGEN KINETICS DATA AT =1200°C FOR OXIDE + ALPHA (Xi) LAYER THICKNESS



#### **SUMMARY**

#### Steam Oxidation Kinetics Studies: BWR Zircaloy-2

- Weight gain data at =1200°C for unirradiated/irradiated Zry-2 agrees quite well ( $\overline{6}$ %) with Cathcart-Pawel (CP) model
- Xi (oxide+ $\alpha$ ) thickness data larger (=27%) than CP model Irradiated cladding has highly irregular  $\alpha$ - $\beta$  interface

#### LOCA Integral Tests

- 1st high burnup BWR test scheduled for April 2002
   Aggressive test with 10-minute hold time at PCT = 1204°C
   2-5 additional BWR tests with varying test-times/ECRs
- PWR LOCA tests will follow with 6-12 samples taken from GS#2 (50-μm OD oxide) and GS#4 (100-μm OD oxide)